



Dissolved zinc and copper retention from stormwater runoff in ordinary portland cement pervious concrete



Liv Haselbach*, Cara Poor, Jerin Tilson

Civil and Environmental Engineering, Washington State University, 405 Spokane Street, Pullman, WA 99164-2910, United States

HIGHLIGHTS

- Pervious concrete removes significant amounts of dissolved zinc from stormwater.
- Pervious concrete removes significant amounts of dissolved copper from stormwater.
- Pervious concrete can perform well for copper and zinc removal over years of use.

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ABSTRACT

This research investigated whether pervious concrete is effective in removing dissolved zinc and copper contaminants from stormwater runoff. Pervious concrete cylinders were loaded in the laboratory with simulated runoff, and influent and effluent concentrations and volumes measured. In one set of tests, stormwater was dispensed at typical concentrations (100 $\mu\text{g/L}$ Zn and 20 $\mu\text{g/L}$ Cu) simulating ten consecutive storm events of 12 mm (0.5 in.) of rain to test initial removal performance. Another set of cylinders was loaded at forty times the typical concentration to simulate accelerated longterm metal loading indicative of several years of typical use. Cylinders were removed after 15 and 24 accelerated events respectively for further typical concentration performance testing. Performance for the typical concentration tests both pre and post accelerated loading were calculated as a percent metals retained and also as a percent decrease in effluent versus influent concentration. The pre accelerated metal loading results indicate averages of 93% and 91% retained, and 90% and 87% concentration decreases for Zn and Cu respectively. Similar percentages were retained post accelerated metal loadings with slightly lower averages for copper after 24 accelerated loading events, although this was probably due to a higher infiltration rate. Removals were seen to be effected by both physical (water trapping) and by chemical (sorption and complexation) processes.

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1. Introduction

Urbanization may have negative environmental impacts as stormwater runoff from paved surfaces and roofs increases, possibly causing erosion and carrying additional stormwater and related pollutants to water bodies. Stormwater runoff may contain elevated levels of zinc and copper which may pose a significant threat if discharged directly to many aquatic environments [1–4]. Permeable pavements allow rainwater to infiltrate into the ground, reducing many of the potential stormwater impacts of paved surfaces. Some of these permeable pavements may also serve an additional use for stormwater quality treatment.

Some studies have indicated that permeable pavements are effective in removing metals from stormwater, especially

particulate bound metals. Most of these studies focused on full permeable pavement systems, consisting of the pavement layer (pervious concrete, porous asphalt or permeable pavers), the aggregate storage bed below and other attributes of the system such as the below grade soils. Mechanisms may have included filtration of particulate bound metals and sorption to soils and other particles in the systems [5,6]. This study is based on the hypothesis that the pervious concrete layer in a pervious concrete pavement system would effectively remove dissolved zinc and copper due to the chemical and physical characteristics of the concrete such as the high pH, the carbonate species, the high surface areas for sorption in the tortuous channels and physical capture of water in the interconnected pores with subsequent evaporation and precipitation.

When ordinary portland cement (OPC) is used to make concrete, the compounds that result include many hydroxide species such as calcium hydroxide. In the tortuous interconnected flow channels throughout pervious concrete, there are many carbonate

* Corresponding author. Tel.: +1 509 335 4874; fax: +1 509 335 7632.
E-mail address: haselbach@wsu.edu (L. Haselbach).

species, in addition to the hydroxide species, on the surface due to carbonation from the carbon dioxide in the atmosphere. In fact, X-ray photoelectron spectroscopy (XPS) analyses have indicated that on cement surfaces there may be up to three carbon atoms for every calcium ion [7]. Dissolved metals are likely removed from solution while passing through these tortuous flow channels within a pervious concrete layer due to complexation and solids formation with the hydroxides (OH^-) and carbonates (CO_3^{2-}) that are attached to calcium in the concrete. In aqueous systems, many complexes form between carbonate and hydroxide ligands, and metals such as calcium, zinc and copper. Formation of these complexes and solids with the dissolved copper and zinc is also enhanced at higher pH which is typically found in OPC concrete. These complexes can also form on surface species of the carbonates and hydroxides [8]. Both dissolved copper and zinc form complexes with water in aqueous solutions very rapidly. These complexes then tend to sorb more strongly to surfaces than the free ions would. Both copper and zinc in sorbed complexes of hydroxides and carbonates are then preferred in pH ranges above 6.5, with the carbonate complexes the predominant species until pH 9.5 [9]. Thus, there is a high affinity for surface complexation of the dissolved zinc and copper ions with both ligands. After complexation during a storm event, there is then the opportunity for diffusion into the microporous structure of the interior pervious concrete matrix. As in any ordinary portland cement concrete specimen exposed to the atmosphere, beyond the highly carbonated surfaces, the concrete will be less carbonated and there will be more hydroxide species remaining [10]. These interior sites may also form many complexes with the zinc and copper ions, with the potential for formation of the solid complex species too. The logarithm of the stability constants for the formation of the solid hydroxide species for the three metals are 5.19, 19.3 and 16 for calcium, copper and zinc respectively, indicating a higher affinity for formation of the copper and zinc hydroxide solids than the calcium. The logarithm of the stability constants for the formation of the solid carbonate species for the three metals are 8.2, 9.6 and 10 for calcium, copper and zinc respectively, again indicating a higher affinity for copper and zinc than calcium. In addition to the solid phases, the stability constants for the various complexations generally tend to favor zinc and copper complexation over calcium [8].

There are some previous studies which support the hypothesis that OPC concrete may have these sorption capabilities. Fach and Geiger [11] developed metal adsorption equilibrium curves (isotherms) for the concrete in concrete pavers by pulverizing them and exposing crushed samples to aqueous dissolved metal solutions. Substantial amounts of the metals did complex, but, these studies had much more exposed surfaces than pervious concrete [11]. Dyer et al. [12] found that significant amounts of metal hydroxides precipitate on the concrete surface and within the concrete in a study on the impact of soil contaminated with zinc, lead and copper on freshly placed concrete [12].

Due to the large amounts of surface areas in the channels in pervious concrete, the likely processes for pervious concrete sorption of dissolved copper and zinc are rapid surface adsorption forming various complexation species during a storm event, and then diffusion and additional complexation species development in the interior of the concrete matrix as pervious concrete may be viewed more as a fractured porous media, than an unfractured porous media, and diffusion is important in the fractures [13,14]. This three-step process would then allow for partial renewal of the complexation sites in the flow channels, i.e. loss of the metal cations through diffusion into interior pores, prior to subsequent storm events. Due to the high levels of the carbonate and hydroxide species in the pervious concrete, and the relatively low concentration of dissolved zinc and copper in typical runoff, it was

predicted that pervious concrete would not to saturation and lose its sorptive capabilities even after many years of use.

The study reported hereupon was conducted in three parts using laboratory testing to simulate field conditions. The intention of the first set of tests (Part 1) was to examine the initial removal of dissolved zinc and copper species by new placements of pervious concrete from simulated stormwater runoff with typical dissolved metal concentrations. The intention of the second part (Part 2) was to simulate longer term loading by accelerating many years of metal sorption by loading specimens with very concentrated solutions of these two metals. In this second portion removal efficiencies were also determined. Although these efficiencies are not realistic of in situ conditions, they are indicative of metal mass loadings and therefore years of service. The intention of the third part (Part 3) was to examine the effectiveness of the pervious concrete to remove typical concentrations of dissolved copper and zinc after the accelerated longer term loading, or perhaps to even begin leaching of the metals.

2. Materials and methods

2.1. Pervious concrete specimens

Pervious concrete cylinders were prepared in December 2008 using a mix of ordinary portland cement, narrowly graded number eight sized basalt aggregate (nominal diameter of 9–10 mm), and water, in mass percentages of approximately 19% cement, 76% aggregate and 5% water respectively. The materials were combined in a concrete mixer and then scooped into cylindrical molds and the fresh specimens compacted approximately 10% of the total depth by pressing down on the top with a traditional concrete cylinder. The specimens were then covered and left to cure for at least seven days prior to removal from the molds. The cylinders were all 102 mm (4 in.) in diameter and ranged in volume from 0.00147 to 0.00151 m^3 (90–92 in^3). These pervious concrete cylinders were previously exposed to tap and deionized water for pH testing in 2009–2010 and then left open to the atmosphere in the laboratory. The previous pH tests performed included both flow through tests and also tests where the cylinders were submerged in water for 30 min and the bath water tested for pH [15]. This allowed for carbonation to develop in the flow channels, typical of in situ conditions. The porosity of each cylinder was measured in February 2012 using a modified ASTM C1754 [16]. All cylinders were then flushed with tap water before testing to remove miscellaneous material. They were chosen from a larger group of specimens due to their similar porosities which minimized experimental variability from differing porosities. All the cylinders were prepared at the same time in the same batch of concrete using the same cement and aggregate source, further decreasing experimental variability. All the cylinders were wrapped on their sides with shrinkwrap to simulate column setups, allowing for water to be poured on the tops and exfiltrated through the bottoms without horizontal losses along the depths. The shrinkwrap decreased side channelization that might have occurred if placed in hard plastic columns.

Part 1 (initial typical removal rates) was performed in triplicate on three cylinders simultaneously. Part 2 (longterm loading) was performed on the remaining six cylinders simultaneously. Part 3 was performed on two of the cylinders from Part 2 after a set number of Part 2 tests to evaluate performance after extensive metal loading, while the remaining cylinders from Part 2 continued with longer term loading events. All the cylinders and the testing that they received are summarized in Table 1.

2.2. Simulated rainwater, stormwater and influent/effluent sample preparation

Simulated stormwater was prepared in two steps. First, simulated rainwater without metal concentrations was prepared and then the metals were added from a highly concentrated stock solution of zinc chloride and cupric chloride dihydrate to achieve the various metal concentrations. The simulated rainwater was made based on the chemical makeup of rainwater in Southeast Washington State as previously analyzed at Washington State University. Sodium chloride, calcium carbonate, potassium nitrate, sodium bicarbonate, and potassium bicarbonate were added to deionized water to achieve 10^{-5} mol/L concentrations of Na^+ , K^+ , Ca^{2+} , Cl^- , NO_3^- , and total inorganic carbon (TIC) of 4.63, 4.17, 0.30, 4.23, 3.88, and 1.00 respectively. The basin of simulated rainwater was left exposed to the atmosphere in the laboratory and the pH subsequently measured at 6.4. This then resulted in estimated concentrations for Na^+ , K^+ , Ca^{2+} , Cl^- , NO_3^- , and TIC as carbon in $\mu\text{g/L}$ of 1070, 1630, 120, 1500, 2410 and 270 respectively, with a little less than half the TIC in the form of dissolved carbon dioxide (H_2CO_3) and a little more than half as the bicarbonate ion, with negligible carbonate species [9].

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